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HEAVY METAL LOAD OF SOILS AROUND A WASTE ROCK DUMP IN THE MÁTRA MTS., HUNGARY

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ABSTRACT

Soils around a waste rock dump were studied in the Recsk-Lahóca sulphide ore mining area (NE Mátra Mts., N Hungary). Three soil profiles were examined with a 50 m lag along a slope of the dump. Heavy metals (Cu, Co, Fe, Mn, Ni, Zn, Pb) were measured as well as Al, pH(H_2O) and C_{org}. Soil chemical and statistical analyses show that Zn and Cu accumulate in the topsoil due to resedimentation from the dump material. Fe, Mn, Ni and Co are of lithogene origin in each profile. In sections close to the dump a significant increase of pH can be observed in the topsoil (up to 7.5) due probably to calcite dissolution. Calcite, as a rock forming constituent of the ore-bearing limestone, is present only in the dump material, because the study area consists mainly of volcanic rocks. Although pH is regionally low (around 5.5) in the study area, and the common clay mineral in the soils is kaolinite, the local control of pH is responsible for hindering mobility of the possibly pollutant metals both vertically and horizontally. Due to this effect the dump studied is no risk to its environment.

Key words: waste rock dump, soil pollution, heavy metal mobility, carbonate buffer.

INTRODUCTION

In the Mátra Mts. in Northern Hungary several inactive dumps of the earlier mining activity can be found. Until the last decade sulphide ore minerals, chalcopyrite, enargite, sphalerite, galena as well as precious metals were extracted from underground mines. The dump material still contains a significant amount of heavy metals and may have an important role in heavy metal pollution of the soils nearby (e.g. Korte et al., 1976; Merrington and Alloway, 1994). At the NE foot of the mountains a 20 km^2 large drainage basin was examined (Fig. 1). In the framework of a complex geoecological evaluation (Mezősi and Rakonczai, 1997) among several problems (natural vegetation, drainage system, soil erosion, etc.) also chemical and physical features of the soil was investigated and mapped. Using data of 150 soil profiles, these studies (Farsang, 1996a) showed that the major element as well as the heavy element distribution of the soils over the study area exhibit a natural signal and essentially is free from significant external pollution. No small-scale examination has, however, been carried out yet, there is no information about soil chemistry around the heavy element accumulating dumps. The aim of the present study is to examine the change of the heavy metal concentration in soil profiles moving away from one dump, as well as to test how well soil could buffer occasional pollution.

GEOGRAPHY, GEOLOGY, PEDOLOGY

The study area lies at the NE foot of the Mátra Mts. at 200-800 m above see level (Fig. 1) with increasing average elevation to the west. The 20 km^2 small catchment area is surrounded by peaks in all directions defining the drainage divide, its main stream flows out the area on the eastern end. The average relief is in the range of 95-150 m/km^2. The area is characterized by a temperate climate with 650-750 mm/year rainfalls and 8.3-8.5 °C annual average temperature (Somogyi, 1990). There is only one small village in the basin, Bodony.

Geologically, Paleogene volcanic and sedimentary rocks dominate on the surface. The most common rock types over the whole study area are Eocene andesite, dacite, rhyolite as well as their tuffaceous equivalents with a small amount of

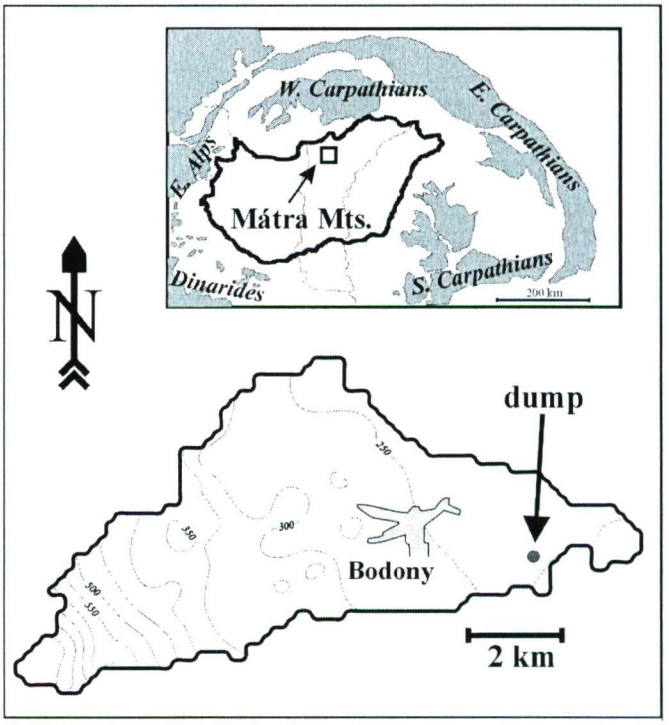

Fig. 1. Topographic sketch map of the studied catchment area. Inset: position of the Mátra Mts. within the framework of the Alp-Carpathian system.

Oligocene clayey sediments in the deeper sub-basins (Báldi, 1983). At places Miocene pyroxene andesite lava flows cover the underlying strato-volcanic complex. Ore minerals of epithermal, porphyry, vein and skarn types are associated with the Eocene andesite. Also Triassic limestone, below the Paleogene formations contains sulphide ore bodies of Cu, Zn, (Cd) and Pb skarn and vein types. Carbonate rocks do not crop out in the area; along the small streams and creeks Pleistocene and Holocene fluvial sediments cover the older formations.

The characteristic soils of the whole catchment area are different subtypes of brown earth (Cambisols and Luvisols). In small patches also Fluvisols, Leptosols (according to the FAO classification; FAO, 1974) occur, following the lithological and/or topographical diversity. Texturally the soils are clayey with only a little regional variation. Physical as well as chemical characters of the soils were mapped based on 150 sampling points with random spatial distribution (Farsang, 1996a, b). Organic material content defines a background of around 2% with a sharp contrast in the forest areas (5%). pH follows the lithological conditions being close to neutral above the loamy sediments and varying around 5 in the parts covered by the volcanic rocks (Fig. 2). The geological background essentially determines large-scale variation of heavy metal content of the soils as well. All studied elements show maximum values at the most elevated areas, where the large relief energy results in thin, lithomorphic soil profiles. Here the volcanic rocks at the surface act as the most important source causing high concentration values. In this paper we recall topsoil (0-20 cm depth) concentration maps of Co and Pb as examples (Fig. 3A, B). On these maps one can follow the co-variation tendency of heavy metal contents with pH and topography. The highest regions consist of volcanic rocks and consequently serve low pH and large heavy element concentration values.

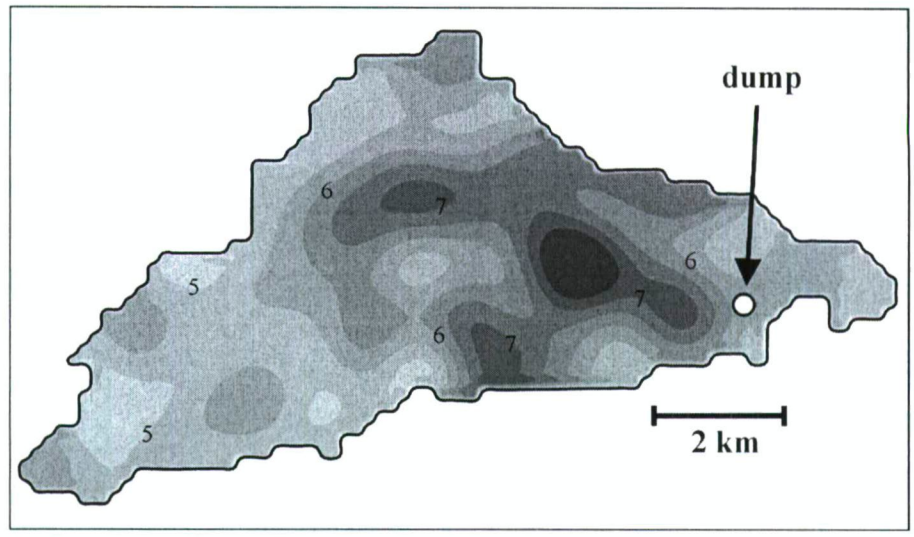

Fig. 2. Spatial pH distribution of the studied catchment area follows lithological variation being around neutral above the loamy sediments and 5-6 on volcanic bedrocks.

The waste rock dump studied can be found at the SE part of the catchment area (Fig. 1). It is a 15 m high hill with a diameter of ~60-70 m at the base (Fig. 4A). The bedrock is andesite, the typical brown earth soil profiles around it are 70-80 cm thick and are covered by a thin layer (up to 3 cm) of the resedimented dump material (Fig. 4B).

Methods

During the geoecological examination of the area, soil samples were collected from 150 points, which represent the topsoil (0-20 cm) and 40 cm depths, respectively. Additional soil profiles represent the surroundings of the potential sources of pollution, including the dump in question. One profile was made at the foot of the dump (P1), and two others away from with a 50 m lag downstream along the northern slope (P2, P3). Each profile was sampled down to the bedrock at every 5 cm.

Following extraction in aqua regia (in a Gerhardt-Kjeldaltherm block), concentrations of Cu, Co, Fe, Mn, Ni, Zn, Cd, Pb and Al were measured using a Perkin Elmer 3010 AAS machine at the University of Tübingen (Germany), following the instructions of Beck et al. (1995). Analytical errors are as listed: Cu: 4%, Co: 3%, Fe: 4%, Mn: 2%, Ni: 5%, Zn: 7%, Cd: 4%, Pb: 8%, Al: 8%. Also pH(H_2O) was measured being a key variable governing heavy metal mobility (Maskall et al., 1995; Gäbler and Schneider, 2000).

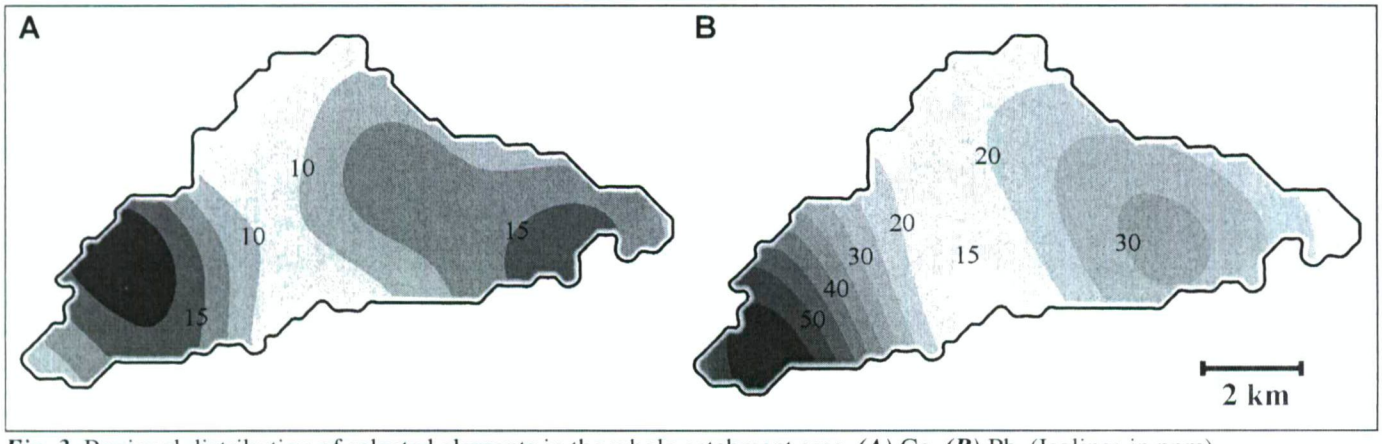

Fig. 3. Regional distribution of selected elements in the whole catchment area. *(A)* Co, *(B)* Pb. (Isolines in ppm).

The main mineral components of the soil samples were determined by standard XRD methods for different size fractions (0-2 μm, 2-5 μm, 5-10 μm, 10-20 μm, >20 μm) along all profiles. Clay minerals were determined by swelling with glycole in settled preparates. Organic matter (C_{org}) was determined in a Philips PU8675 VISible spectrophotometer following oxidation with $K_2Cr_2O_7$.

In order to specify how different heavy elements do accumulate, XRF measurement on different size fractions (0-2 μm, 2-5 μm, 5-10 μm, 10-20 μm, >20 μm) of the soil profiles P1 and P3 were carried out. Because the amount of the material was insufficient for a standard XRF measurement, we used the energy dispersive spectra. These were compared with standard 10, 50 and 100 ppm spectra for estimating concentrations.

Results

Heavy metal content of the soil profiles

Heavy metal concentration as well as Al, pH(H_2O) and C_{org} data from the 3 soil profiles are given in Table 1. Typical concentrations for each element are in the range common for brown earths (Kloke, 1980), they do not exceed the health limit according to the current Hungarian directives. Only Cu in the topsoil (0-2 cm) of P1 is rather close to it (93 ppm). Vertical distributions of the studied elements exhibit characteristic trends following their common geochemical features. Siderophile elements (Fe, Mn, Co, Ni), which accumulate in the silicate phases of the bedrock reach their maxima in the B horizon at around 40 cm depth. Chalcophile elements (Zn, Cu, Pb) show different kinds of trends (Fig. 5); Zn is decreasing downwards in all cases similarly to Cu in the cases of P1 and P2. Cu is equally low along the whole P3 profile. Pb has a random distribution along the profiles and shows no significant tendency to accumulate.

In the case of the P3 profile, pH does not alter along the section (5.2-5.5) and has similar characteristics to common brown earth soils in the study area. In the two other cases, however, pH exhibits a disturbed trend with a significant increase towards the surface (5.5 → 7.5, Fig. 5).

Qualitative XRD measurements suggest that above 2 μm quartz and K-

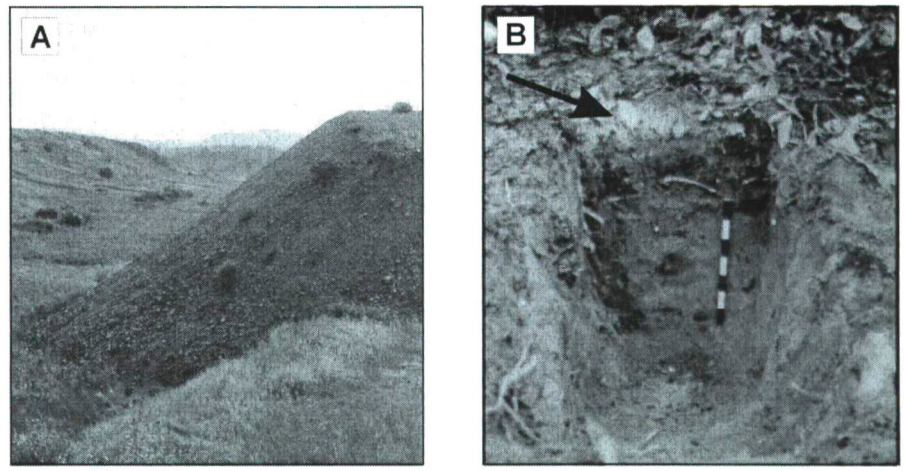

Fig. 4. ***(A)*** Photograph of the studied ore dump looking to the East. Northern slope of the dump was sampled. ***(B)*** The P1 soil profile. Arrow points to the resedimented dump material at the top of the section.

Table 1. Heavy metal and pH data in the soil profiles studied (depth in cm, concentrations in ppm)

	Depth	pH	C_{org}	Zn	Cu	Pb	Co	Ni	Fe	Mn
	2	7.6		80.0	43.0	32.1	15.8	13.0	21350	1063
	5	7.6		80.0	45.0	25.0	15.9	14.8	22310	875
	10	7.6		67.5	37.5	37.5	15.5	14.4	22537	975
	15	7.4		62.5	29.5	36.2	13.7	15.3	21612	1200
	20	7.3		63.8	25.7	19.7	17.4	16.0	22932	1375
	25	7.2		60.0	18.8	25.0	18.2	14.7	20475	1275
	30	6.8		50.0	22.5	35.3	23.3	11.9	22168	1650
P1	35	6.0		53.8	24.0	26.6	22.1	10.3	20783	1400
	40	6.2		52.5	22.2	29.5	15.2	10.1	21400	1062
	45	5.9		48.8	17.5	21.6	16.9	11.4	19806	1012
	50	6.1		52.5	24.7	33.7	13.4	11.6	20993	900
	55	6.3		47.5	27.3	25.3	18.4	13.1	21985	1037
	60	6.3		55.0	28.0	20.0	17.9	16.0	23867	887
	65	6.6		52.5	28.0	14.4	18.4	10.8	23798	825
	70	6.7		55.0	35.7	20.2	17.4	17.6	25595	812
	75	6.7		55.0	32.2	18.7	14.6	12.0	24797	675
	2	7.6		141.3	93.3	33.7	4.7	10.5	12706	112
	5	7.8		58.8	44.0	17.5	12.6	8.3	18987	662
	10	7.7		65.0	36.5	41.3	13.9	11.8	22740	812
	15	7.4		62.5	22.0	38.0	13.5	13.5	22150	1037
	20	7.2		55.0	18.5	23.7	14.9	12.4	20337	1000
	25	7.0		61.3	20.6	32.7	19.4	14.0	23533	1250
	30	6.2		58.8	22.0	39.0	14.9	13.5	23837	1062
P2	35	5.7		55.0	23.7	29.5	16.9	12.1	23450	1187
	40	5.5		52.5	24.3	9.5	18.5	12.2	22672	1275
	45	5.6		50.0	25.1	30.0	23.1	16.8	23626	1275
	50	5.7		80.0	40.8	37.5	24.2	17.7	39125	1437
	55	5.7		52.5	23.8	15.0	14.3	12.1	21568	762
	60	6.1		61.3	26.3	26.2	21.9	11.6	23482	937
	65	6.0		50.0	26.8	33.5	14.3	13.3	23625	787
	70	5.6		52.5	24.8	17.5	22.2	12.4	24303	900
	2	5.3		67.5	20.0	32.0	6.9	13.7	20495	1187
	5	5.3		73.8	20.6	42.5	13.2	14.7	22036	1175
	10	5.4		66.3	20.9	21.3	12.0	19.1	23358	1137
	15	5.3		58.8	17.3	32.9	9.2	11.6	20172	950
	20	5.3		61.3	18.8	35.0	13.4	13.6	22400	1087
P3	25	5.3		60.0	18.8	27.5	14.1	15.1	22662	1487
	30	5.3		53.8	18.8	40.0	16.5	15.5	25075	1412
	35	5.3		53.8	21.9	35.4	25.5	14.5	26975	1937
	40	5.3		50.0	17.5	30.0	17.5	13.5	21781	1250
	45	5.5		48.8	17.5	30.7	22.2	11.6	25850	1375
	50	5.5		48.8	20.3	39.3	17.1	11.9	23100	1025
	55	5.2		60.0	20.0	23.7	21.4	13.5	29187	1262

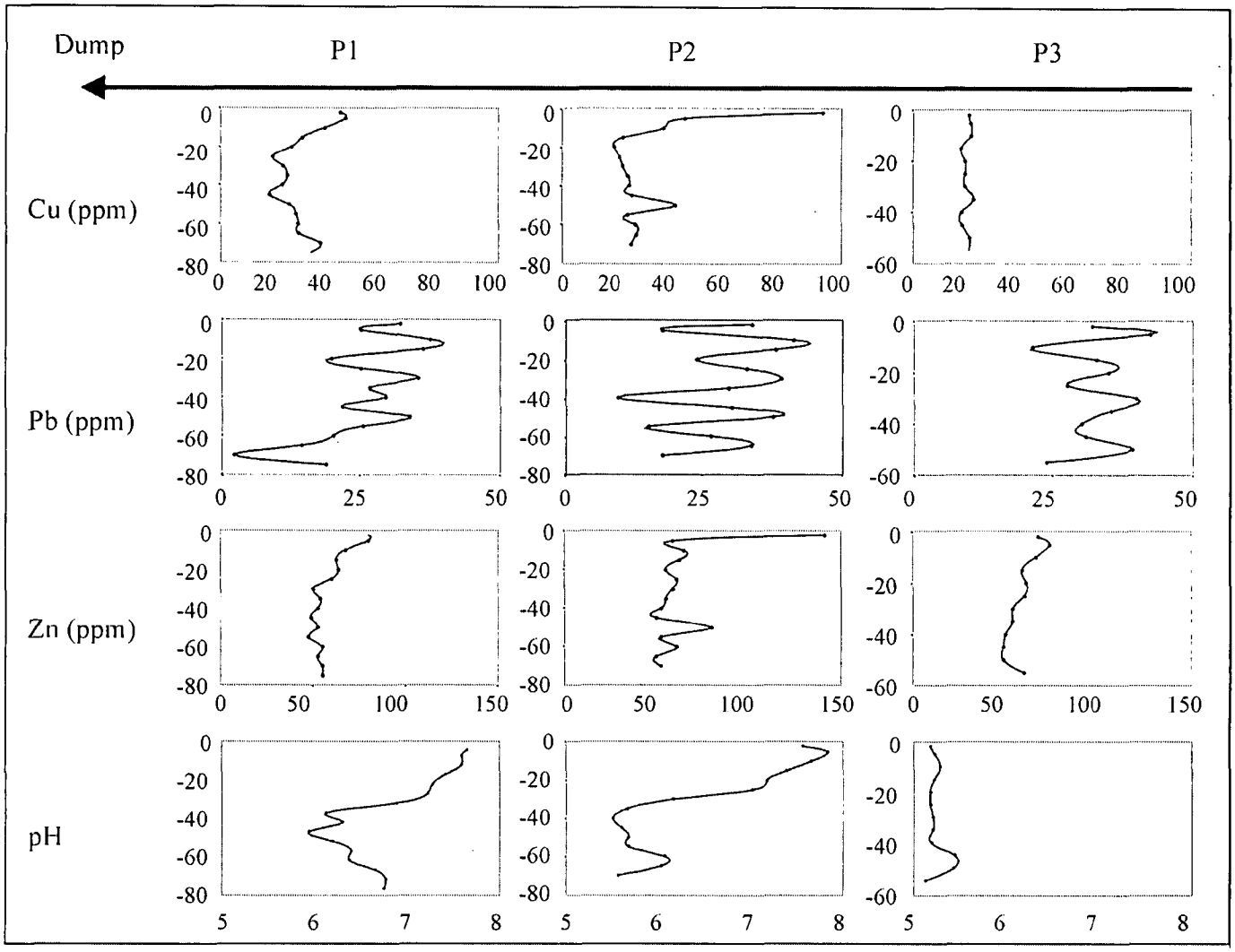

Fig. 5. Vertical variation of Zn, Pb, Cu and pH along the three profiles studied.

feldspar are the dominating phases, while below 2 μm phyllosilicates, over all kaolinite occurs as well. There is only a little detectable variation in the mineralogical composition among the three soil profiles; illite/smectite and chlorite appear exclusively in the P3 profile (Fig. 6). Calcite is present in P1 down to 40 cm, and also in P2 in the upper 20 cm. There is no detectable calcite in P3.

According to the semiquantitative XRF spectra Fe, Mn, Co and Ni reach their peak concentration in the >20 μm grain size fraction, while Cu, Zn and Pb are highest in the 5-10 μm interval.

Mathematical evaluation

In order to be able to compare the vertical variation of the metals and pH quantitatively, geostatistical calculations were made. Concentrations of most elements exhibit a clear dependence on depth as it is shown by their diverse accumulation behaviour. That is, why

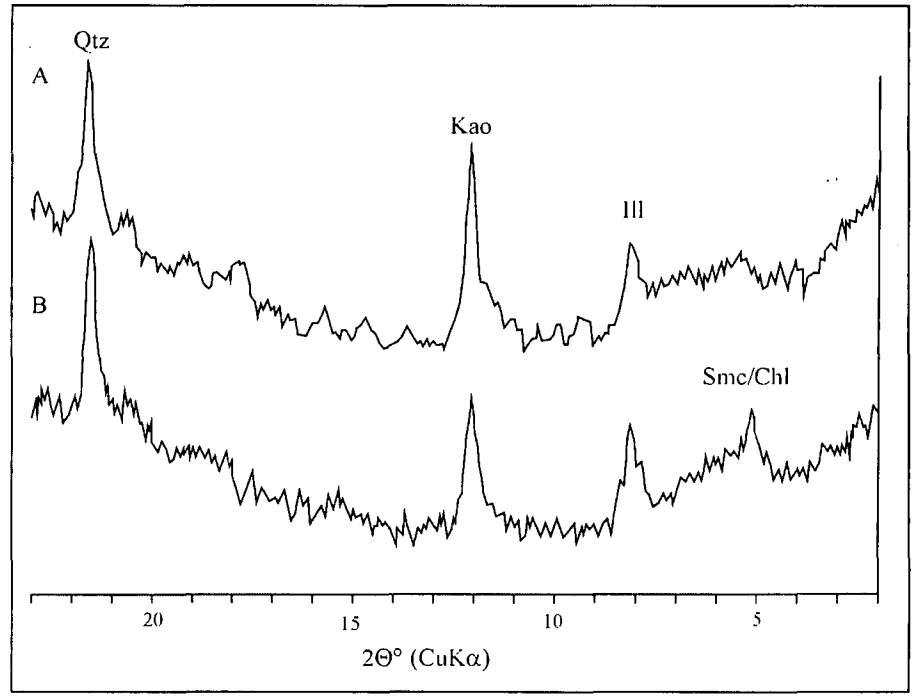

Fig. 6. Selected XRD spectra of settled soil preparates for a) P2, 45-50 cm; b) P3, 45-50 cm

Table 2. The factor score matrix of the three profiles. Significant values are highlighted.

	P1 profile			P2 profile			P3 profile		
Element	factor1	factor2	factor3	factor1	factor2	factor3	factor1	factor2	factor3
Zn	**.88**	-.13	-.22	-.21	**.87**	-.15	-.44	**.85**	.11
Cu	**.71**	.28	-.39	-.38	**.81**	-.29	.44	**.68**	.35
Pb	.35	**-.70**	.00	.54	**.69**	.29	.01	-.19	**.89**
Co	-.24	.01	**.90**	**.66**	-.59	-.14	**.92**	-.29	-.01
Ni	**.65**	.43	.01	**.90**	.01	-.15	.13	**.75**	-.19
Fe	.14	**.94**	-.11	**.89**	-.19	-.25	**.87**	.01	-.21
Mn	.01	-.46	**.83**	**.76**	-.54	-13	**.82**	.13	.12
Al	.34	**.71**	-.40	**.87**	-.22	.01	-.34	**.69**	-.01
pH	**.95**	.01	-.01	-.37	**.60**	.50	.01	-.49	.28
C_{org}	**.76**	-.51	-.04	-.14	-.15	**.84**	**-.62**	.41	.46

prior to variography (e.g. Cressie, 1991), trend functions had to be subtracted. In several cases (Fe, Co, Zn) linear trends were chosen, while in others (Ni, Cu, pH, Mn) a second power function fitted the best. Semivariogram for each variable is of a simple Gaussian type with ranges varying between 15 and 35 cm. Calcophile metals, which accumulate in the topsoil have similar range values for each profile (Zn ~ 23 cm, Cu ~ 21 cm, Ni ~ 18 cm), that is they have an influence down to around 18-23 cm depth. Pb and Mn are rather immobile (Pb ~ 15 cm, Mn ~ 14 cm) opposite to Fe (~ 27 cm) and Co (~ 26 cm). The largest vertical range value is typical for pH (~ 35 cm) in each case.

To emphasize the relationship between the behaviour of different variables, factor analysis (principal component method with varimax rotation) was performed independently for the three profiles. Results are collected in Table 2, where elements with correlation coefficient > 0.6 are pointed out. Calculations resulted in 3 factors in each case suggesting that groups of certain elements have a tendency to change together. Mn, Co and Fe, another one by Cu and Zn can define a group while Al, Pb and Ni alter their positions. Also the role of pH changes from P1 to P3; while for P1 pH has as high as r=0.9 correlation coefficient with Zn and Cu, it has no effect on the elements behaviour in the latter case (r<0.3).

Discussion

Comparison of the profiles

Based on the soil chemical and mineralogical data as well as the statistical results, the laws of the spatial distribution of the heavy metals around the studied dump in the Mátra Mts. can be understood. The three profiles represent different stages of the evolution. While P1 is under the strong influence of the dump material, P3 is almost identical with soils characteristic in other parts of the study area. In this latter case neither positive pH anomaly can be recognized, nor significant chalcophile element accumulation is present in the topsoil. All these values along the whole section are typical for brown earths of the study area. Elements that concentrate in the andesitic bedrock (Fe, Mn, Co) exhibit a clear increasing trend downwards, suggesting lithogene origin in the whole soil profile. The common origin of these elements is also confirmed by the factor analysis. The results show that in case of P3, the „lithogene" factor (Fe, Mn, Co, -C_{org}) has the largest eigen value, suggesting that it determines the chemical nature of the soil. The behaviour of the commonly most conservative Pb is independent of the other chalcophile elements, Cu, Zn and Ni, which have a common peak together with Al; they possibly are adsorbed on clay mineral surfaces. Although, in the whole study area kaolinite dominates the clay mineral fraction, in case of P3 also illite/smectite occurs (c.f. Fig. 6) and could serve as efficient trap for heavy metals because of its high adsorptive capacity (Allard et al., 1991; Lothenbach et al, 1998).

P1 profile, which represents the soil of the dump itself, shows different characteristics. There is a significant accumulation in the topsoil of metals, which redeposit from the dump material (Fig. 4B). Based on the results of the factor analysis, the most determining group of elements is the (Cu, Zn, Ni) concerning the whole profile. In this case also pH varies together with these metals with a very strong positive correlation coefficient. As decomposition of the sulphide ore minerals would significantly decrease pH, in this case another effect should be taken into account. Dissolution of calcite, which is an important constituent of the dump and is also present in the topsoil of P1 and P2, may be responsible for local increase of pH. Calcite disappears close to the bedrock and also pH reaches its normal value (5.5) confirming the above statement. Results of the variography show that pH has a significantly larger range than any metals studied. Although, there is a significant amount of metal on the surface, and in this case the characteristic clay mineral of the soil is kaolinite, which is unable to adsorb the pollution, the locally increased pH may hinder the mobility of Zn, Cu and Ni. As a consequence, the ranges of the metals in the case of P1 are analogous with those calculated in the case of P3.

P2, which lies between the two previous profiles geographically, shows intermediate features concerning the soil chemistry as well. Here, similarly to P3 the lithogene factor (Fe, Co, Mn) has the largest significance characterizing the soil chemistry. On the other hand, there still is a significant role of calcite redeposition and dissolution resulting in a tight connection between pH and the (Cu, Zn Ni) group. Complex behaviour of this profile proves that pH has a broad influence regime not only vertically (range > 30 cm in each case), but also horizontally.

Presence of carbonate minerals is rather strange in the volcanic rocks dominated surface; strictly speaking, they are pollutants around the dump. In the studied brown earth soils, however, which generally are characterized by low pH values and clay minerals of low cation adsorption capacity (kaolinite), calcite helps to diminish metal mobility through increasing pH locally. Data suggest that basically the pH-barrier balanced by calcite deposition and dissolution is

responsible for the fact that P3 profile 150 m far from the dump may chemically be unpolluted. Dudley et al. (1991) described similar sorption effect between natural calcareous soils and acid mine waste. The process essentially is the natural analogue of the crushed limestone barriers commonly applied to mitigate heavy metal migration via adsorption and metal-carbonate fixation (e.g. Artiole and Fuller, 1979). Behaviour of the studied dump also is in harmony with the observations of Tyler and Olsson (2001) who measured decreasing element mobility when adding extra $CaCO_3$ to different soils.

Regional consequences

To determine the sensibility of the soils to heavy metals in the whole catchment area, a buffer capacity map was constructed for each metal based on the values of pH, C_{org} and mechanical structure. Following the algorithm of Marks et al. (1989) buffer capacity can be coded with an integer between 1 and 5 (for details see Farsang, 1996b). In this calculation only those 150 soil samples, which are not under the influence of any dumps were involved. Maps for Ni, Cu and Cd, respectively are presented on Fig. 7. In its present state, the soils buffer Cu and Pb well in the whole catchment, while in the case of Ni and Zn also areas with lower buffer capacity exist. Areas with average or low buffer capacity are the most common in the case of Cd, also the dump studied is located in a region where Cd is buffered insufficiently. A theoretical model, in which we decrease pH values with 1.0 in each datum point, shows that buffer capacity maps would change significantly due to such a supposed environmental effect. In this case not only Cd, but also Ni and Zn would easily be mobilized making the dump possibly dangerous for its close surrounding. Cu and Pb, on the other hand, would not mobilize even in this extreme situation (Fig. 8).

Nevertheless, as we discussed previously, in addition to regional characteristics, the dump studied has its own mechanism to control the mobility of heavy metals. Because relatively large pH around the dump is set up by calcite, which phase deposits and dissolves from the dump material itself, there is no way to get such an acid soil which would be dangerous.

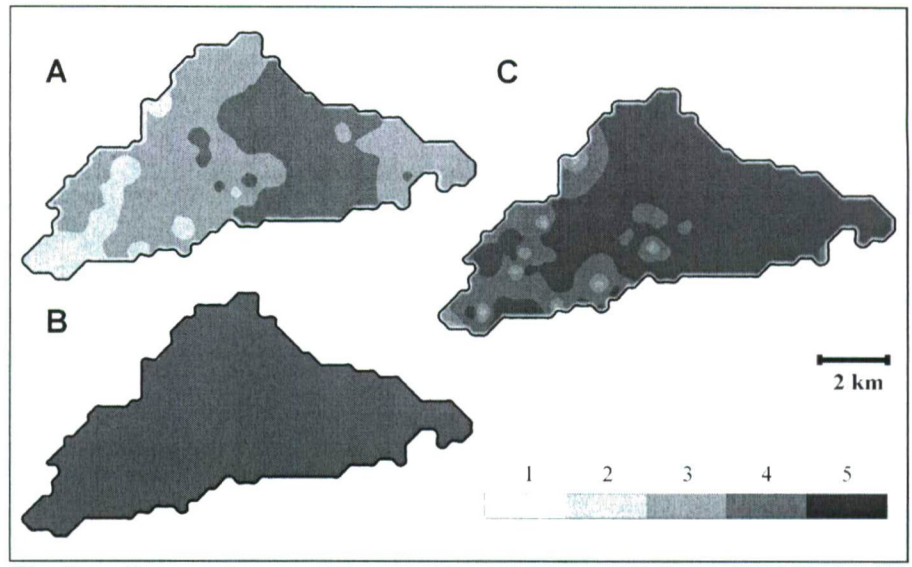

Fig. 7. Buffer capacity maps following the algorithm of Marks et al. (1989) for ***(A)*** Cd, ***(B)*** Cu, ***(C)*** Ni.

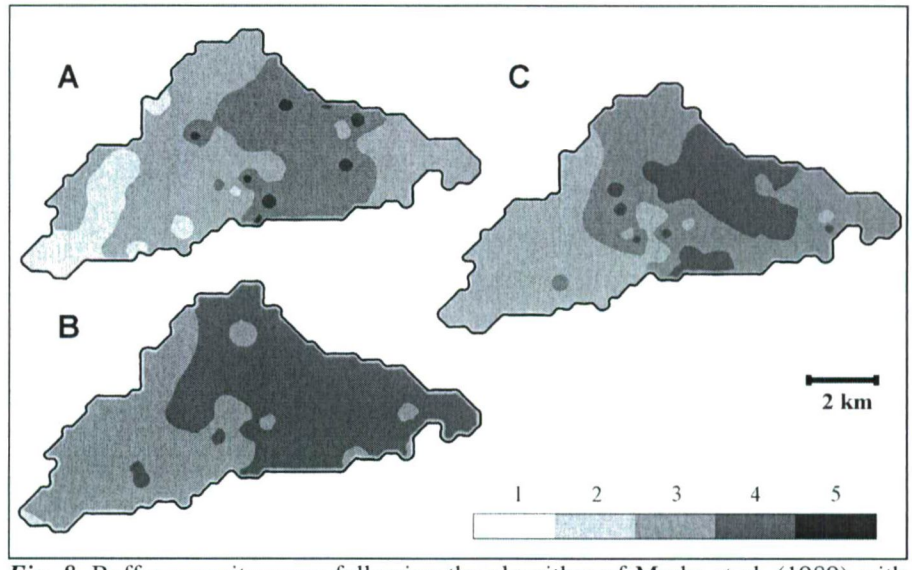

Fig. 8. Buffer capacity maps following the algorithm of Marks et al. (1989) with decreasing pH values by 1.0 for ***(A)*** Cd, ***(B)*** Cu, ***(C)*** Ni.

Conclusions

In conclusion we can state that

- due to resedimentation of the dump material as well as transportation with rain water, Zn and Cu accumulate in the upper horizon of the surrounding soils,
- local deposition and dissolution of calcite increases pH in the close surrounding of the dump,
- the vertical as well as the horizontal movement of Zn, Ni and Cu are hindered due mainly to intermediate pH value. Pb is buffered under each possible conditions well and remains immobile,
- the dump has no environmental risk at its present condition and under possible environmental changes.

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